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ABSTRACT

In support of the Partnership for a New Generation of Vehicles (PNGV), the Idaho National Engineering and Environmental Laboratory (INEEL) has developed novel testing procedures and analytical methodologies to assess the performance of batteries for use in hybrid electric vehicles (HEV's). Tests have been designed for both Power Assist and Dual Mode applications. They include both characterization and cycle life and/or calendar life. At periodic intervals during life testing, a series of Reference Performance Tests are executed to determine changes in the baseline performance of the batteries. Analytical procedures include a battery scaling methodology, the calculation of pulse resistance, pulse power, available energy, and differential capacity, and the modeling of calendar- and cycle-life data. PNGV goals, test procedures, analytical methodologies, and representative results are presented.

INTRODUCTION

Lightweight, compact, high-power energy storage devices are critical enabling technologies for a viable hybrid electric vehicle (HEV) propulsion system. To this end, a cooperative research and development program called the Partnership for a New Generation of Vehicles (PNGV) was formed in 1993 between the Federal Government and the U.S. Council for Automotive Research (USCAR), whose members are DaimlerChrysler, General Motors, and Ford Motor Company (Ref. 1). The U.S. Department of Energy's (DOE) Office of Advanced Automotive Technologies (OAAT) leads the Federal Government's technical and administrative support to PNGV, as well as provides cost-shared funding. Major objectives of the program are to develop technologies for a new generation of HEV's with fuel economies up to three times (80 miles per gallon) the average family sedan. At the same time, these vehicles should maintain performance, size, utility, and cost of ownership and meet federal safety and emissions requirements. [Note: In January 2002 at the Detroit Auto Show, Energy Secretary Spencer Abraham announced that PNGV will be superceded by the formation of a new program between the U.S. Government and the U.S. Council for Automotive Research dubbed FreedomCAR. Its emphasis will be the development of fuel cell-powered vehicles. "The long-term results of this cooperative effort will be cars and truck that are more efficient, cheaper to operate, pollution-free and competitive in the showroom." It is believed that advanced high-power batteries will continue to be a critical component in this new program.]

PNGV ENERGY STORAGE GOALS

PNGV energy storage system performance goals have been developed based on anticipated representative usage and integration with other HEV system requirements. These goals are summarized in Table 1 for both the Power Assist and Dual Mode applications. These goals include pulse discharge and regenerative power, available energy, round-trip efficiency, cold cranking power, cycle and calendar life, weight, volume, voltage and current limits, self discharge, operating temperature range, and cost. To assess battery performance against these PNGV energy storage goals, a cadre of tests and analytical procedures has been developed, and is defined in detail in Reference 2.

Table 1. PNGV energy storage system performance goals

Characteristics	Units	Power Assist	Dual Mode
Pulse discharge power	kW	25 (18-s)	45 (12-s)
Peak regenerative pulse power	kW	30 (2-s) (minimum 50 Wh over 10-s regen total)	35 (10-s) (97-Wh pulse)
Total available energy (over DOD range where power goals are met)	kWh	0.3 (at C_1/1 rate)	1.5 (at 6-kW constant power)
Minimum round-trip energy efficiency	%	90	88
Cold cranking power at -30ºC (three 2-s pulses, 10-s rests between)	kW	5	5
Cycle life, for specified SOC increments	cycles	300,000 Power Assist cycles (7.5 MWh, total)	3,750 Dual Mode cycles (22.5 MWh, total)
Calendar life	years	15	15
Maximum weight	kg	40	100
Maximum volume	l	32	75 (at 165-mm max height)
Operating voltage limits (Note: Maximum current is limited to 217 A at any power level)	Vdc	max $\leq$ 440 min $\geq$ (0.55 × Vmax)	max $\leq$ 440 min $\geq$ (0.5 × Vmax)
Maximum allowable self-discharge rate	Wh/day	50	50
Temperature range: Equipment operation Equipment survival	°C	-30 to +52 -46 to +66	-30 to +52 -46 to +66
Production costs @ 100,000 units/year	$	300	500

PNGV BATTERY PERFORMANCE TESTING

In recent years, the investigation of energy storage devices for HEV's has focused on high-power lithium-ion, lithium-ion polymer, and nickel metal hydride batteries. Prototypical batteries may range from laboratory- and full-size cells, to modules consisting of an ensemble of cells, to full-size PNGV HEV battery systems having electronic and thermal control systems. As PNGV battery suppliers develop new technologies, their batteries are sent to the INEEL for independent and objective testing and evaluation. [Note: Other DOE laboratories that are also supporting DOE's OAAT and PNGV energy storage programs in the areas of abuse tolerance, materials development, low cost packaging and diagnostics, include Argonne National Laboratory (ANL), Brookhaven National Laboratory (BNL), Lawrence Berkeley National Laboratory (LBNL), and Sandia National Laboratories (SNL).]

Prior to starting any test sequence, all equipment is calibrated and all tests are closely controlled at prescribed states-of-charge (SOC), test profiles, and temperatures by using environmental chambers and programmable testers. A measurement and control study of the INEEL Energy Storage Laboratory testers has been completed, and has determined the uncertainty of both measured parameters (i.e., temperature, current, and voltage) and derived parameters (i.e., power, capacity, energy, impedance, efficiency, and self-discharge) (Ref. 3). This information has been utilized to develop precise testing and measurement standards to ensure consistent and objective evaluation over the broad range of products tested in the laboratory.

Following receipt inspection of test articles, a series of characterization tests are performed. These tests include static capacity, hybrid pulse power characterization (HPPC), self-discharge, cold cranking, thermal performance, energy efficiency, electrochemical impedance spectroscopy (EIS), and available energy for Dual Mode batteries (Ref. 2). The static capacity test is a series of at least three complete $C_1/1$ discharges that are repeated until results agree within 2%. This demonstrates charge and discharge stability, helps condition the batteries for further testing, and measures the nominal capacity.

Next, discharge and regen pulse powers are calculated (as described later in this paper) utilizing the low-current Hybrid Pulse Power Characterization (L-HPPC) Test. Figure 1 shows a typical pulse power profile from the HPPC test, which consists of a series of discharge and regen pulses performed at every 10% depth-of-discharge (DOD) increment, with an hour rest at open circuit at each increment to ensure that the battery has electrochemically and thermally equilibrated. Each L-HPPC discharge pulse is performed at the larger of either a 5C current or 25% of the manufacturer's maximum rated current.

A term known as the Battery Size Factor (BSF) is used to scale the remainder of the PNGV power- and energy-based tests. It is either obtained from the battery supplier or it may be calculated from the first series of L-HPPC tests during characterization. The BSF can also be utilized to estimate the unburdened cost, size, and weight of a full-size PNGV HEV battery. Once established, the BSF is generally held constant for the duration of testing. The calculation of the BSF is described in Reference 2.

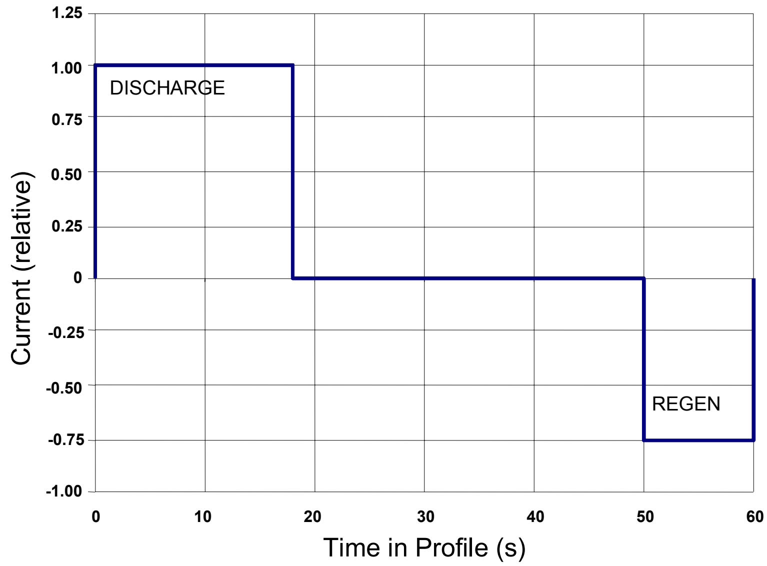

Figure 1. PNGV Hybrid Pulse Power Characterization Profile

Self-discharge is calculated as the difference in energy of a fully-charged battery compared to its energy after sitting at open circuit for seven days. Cold cranking tests measure the battery's ability to provide three consecutive two-second 5-kW pulses at –30°C. Thermal performance is determined by repeating the static capacity and L-HPPC tests at various temperatures. Energy efficiency is determined using a charge-balanced pulse profile and calculating the ratio of watt-hours-output to watt-hours-input. EIS (i.e., full-spectrum complex impedance) measurements are made prior to the start of life testing, and then repeated when life testing is concluded.

Prior to commencing life testing, Reference Performance Tests (RPT's) are executed at 30°C to establish the baseline performance and then are repeated about every 25 days, thereafter. For Power Assist applications, the RPT's consist of a $C_1/1$ Constant-Current Discharge Test and a L-HPPC Test. Whereas, for Dual Mode applications, the RPT's include these two tests plus a 6-kW Constant-Power Available Energy Test.

End-of-testing for all life tests occurs when the device has completed the required time interval or number of cycles, or when it can no longer simultaneously meet the PNGV power and energy goals. For Power Assist applications, the cycle, pulse discharge power, and available energy goals are 300,000 cycles, 25 kW, and 300 Wh, respectively. For Dual Mode these goals are 3,750 cycles, 45 kW and 1500 Wh, respectively. See Table 1.

Calendar-life testing is performed by bringing the battery to a prescribed SOC and temperature and holding at these conditions. Once each day, single discharge and regen pulses are applied from which daily pulse resistances can be calculated.

Life cycling begins by bringing the device to the specified temperature and SOC conditions and performing an Operating Set Point Stability Test to ensure that a stable cycling condition has been established. Figure 2 shows the 25-Wh Power Assist Efficiency and Cycle-Life Profile. It consists of a discharge pulse and a regen pulse with interspersed rest periods. The cumulative length of a single profile is 72 seconds and constitutes one cycle, which is repeated continuously during testing.

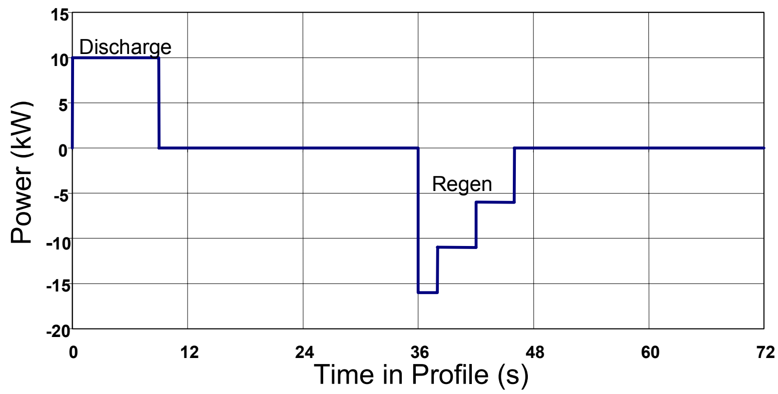

Figure 2. PNGV 25 W-h Power Assist Efficiency and Cycle-Life Test Profile

Figure 3 shows the Dual Mode Cycle-Life Test Profile and the corresponding Net Energy Profile. The power profile is composed of three Dynamic Stress Test (DST) pulse profiles followed by 45 recharge pulse profiles. The three DST profiles are scaled to 36 kW and have a gross discharge of approximately 1500 Wh during this 18-minute sequence. The device is then returned to its initial charge condition using a 72-minute recharge profile sequence followed with a 10-minute rest, for a total duration of 1.667 hours per complete cycle.

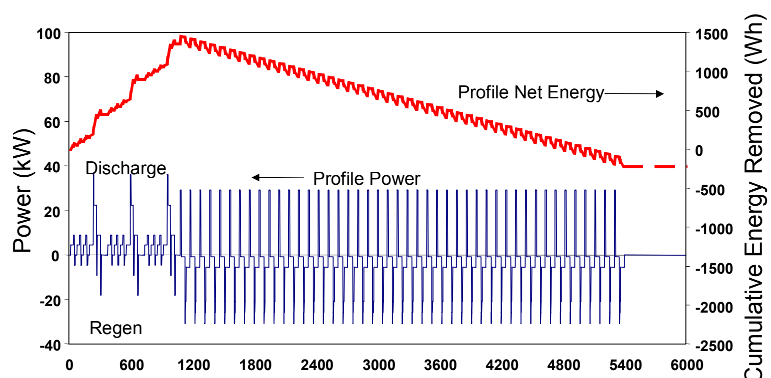

Figure 3. PNGV Dual Mode Cycle-Life Test Profile

Validation of battery performance with respect to the PNGV cycle-life and calendar-life goals at normal HEV operating conditions is a lengthy process. Hence, national laboratories including INEEL, SNL, and ANL have been investigating methodologies to accelerate both calendar-life and cycle-life testing. Typically, these developmental methodologies employ distributing ostensibly identical cells within a test matrix at various SOC's, temperature, and test profiles and then executing the test for prescribed periods of time. As with standard PNGV battery testing, the aging periods are interrupted periodically to execute RPT's from which cell performance as a function of time and matrix variables may be ascertained. These data are then utilized to develop predictive models, typically utilizing an Arrhenius-based approach for temperature dependence, to extrapolate life predictions to normal operating conditions. Reports of INEEL's, SNL's, and ANL's work are found in Reference 4.

ANALYTICAL METHODOLOGIES

Power fade (which is directly related to resistance growth) has been identified as a limiting factor for PNGV HEV batteries. Thus, testing and analytical assessments are largely focused on this parameter. Capacity fade is another key parameter that is tracked during cell testing.

Performance data from full-size, 12-Ah, lithium-ion, Year 2000-configuration Saft HP-12 cells are used as an example to show how PNGV performance parameters are calculated. Characterization testing was begun on these cells at the INEEL in December 2000. They then began cycle-life testing at 25% DOD in February 2001. Two cells each are being tested at 30°C, 40°C, and 50°C and to-date have successfully completed over 190,000 PNGV 25-Wh Power Assist life cycles.

The change in $C_1/1$ capacity with aging for the six cells is shown in Figure 4. As can be seen, the cells initially displayed a very slight increase in capacity with aging, but then began to monotonically decrease after about the third set of RPT's. After 190,000 cycles, the average capacity fade is 9.4% for the 30°C cells, 7.9% for the 40°C cells, and 6.3% for the 50°C cells. Interestingly, over the range tested, the magnitude of the capacity fade decreases with increasing temperature for these cells.

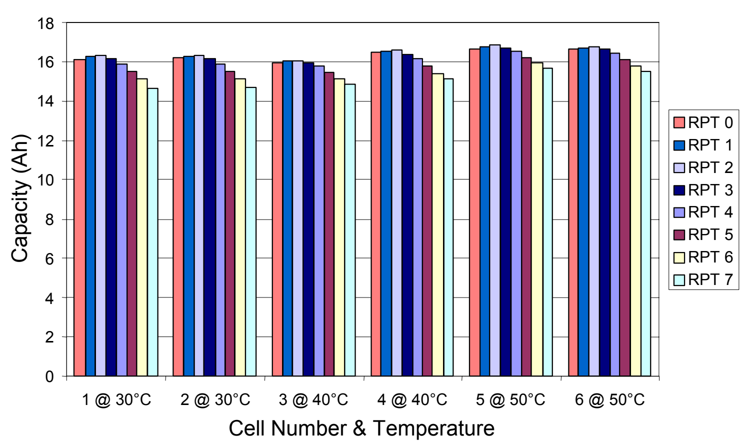

Figure 4. Capacity Summary for Saft HP-12 Li-Ion Cells from Beginning of Testing through 190,000 Cycles

The first step in determining the pulse power capability is to calculate the discharge pulse resistances, $\boldsymbol{R_{dis}}$, and the regen pulse resistance, $\boldsymbol{R_{reg}}$, at each 10% DOD increment from the L-

HPPC test data. Pulse resistance is simply the ratio of the change in the voltage divided by the change in current at specified times during selected pulses. For Power Assist batteries, $\boldsymbol{R_{dis}}$ is calculated from the beginning to 18 seconds into each discharge pulse, and $\boldsymbol{R_{reg}}$ is calculate over the first two seconds of each regen pulse. In contrast for Dual Mode batteries, $\boldsymbol{R_{dis}}$ is calculated over the first 12 seconds of each discharge pulse, and R_{reg} is calculated over the first 10 seconds of each regen pulse. Thus, for the constant-current pulses used in the L-HPPC test, $\boldsymbol{R_{dis}}$ and $\boldsymbol{R_{reg}}$ are given by (Ref. 2)

$$R_{dis} = \Delta V_{dis} / I_{dis}$$

$$R_{reg} = \Delta V_{reg} / I_{reg}$$

where ΔV_{dis} and ΔV_{reg} are the changes in the discharge and regen pulse voltages over the specified time intervals, and $\boldsymbol{I_{dis}}$ and $\boldsymbol{I_{reg}}$ are the corresponding currents. Figure 5 shows the discharge and regen pulse resistance curves and the voltage curve versus DOD for one of the Saft cells at the beginning of testing (solid lines) and after 190,000 cycles (dashed lines) at 30°C. Note that as expected, the resistances increase with aging.

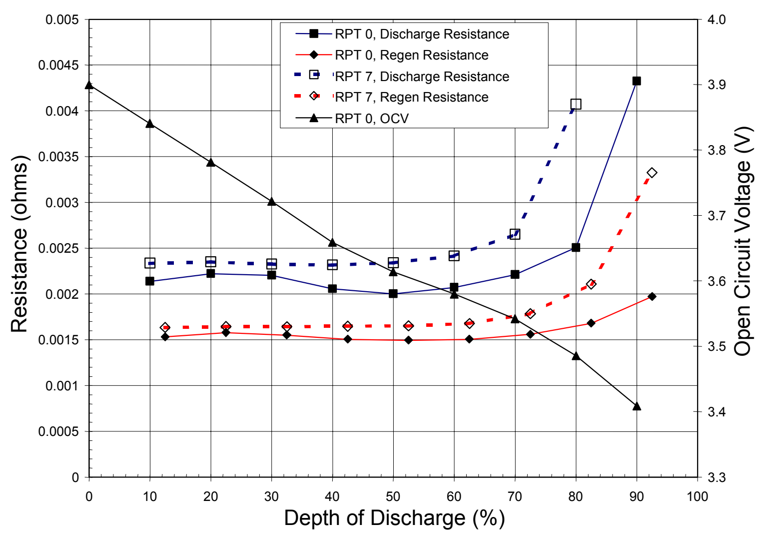

Figure 5. Saft Li-Ion Cell Pulse Resistances and Open Circuit Voltage at Beginning of Testing and after 190,000 Cycles

At each DOD increment, this information is used to calculate the discharge and regen pulse power capabilities. The discharge pulse power capability, $\boldsymbol{P_{dis}}$, and the regen pulse power capability, $\boldsymbol{P_{reg}}$, at each DOD are determined by (Ref. 2)

$$P_{dis} = V_{min} \, (V_{OC} - V_{min}) / R_{dis}$$

$$P_{reg} = V_{max} \, (V_{max} - V_{OC}) / R_{reg}$$

where V_{min} and V_{max} are the manufacturer's specified minimum and maximum allowable voltages and V_{OC} is the open-circuit voltage immediately before each pulse.

Each DOD can also be related to the corresponding amount of energy discharged to that point. Figures 6 shows the corresponding discharge and regen pulse power curves versus energy for the same cell at the same two times in life. For these cells, the BSF was 44.3. That is, it was determined that 44.3 cells would be required to meet the PNGV Beginning-of-Life (BOL) power and energy goals for a full-sized HEV battery. Thus, individual cell values are multiplied by 44.3 to obtain Figure 6. Again as expected, the power capability decreases with cycling, but interestingly for these cells, little or no power fade is observed for the regen pulse power in contrast to the discharge pulse power.

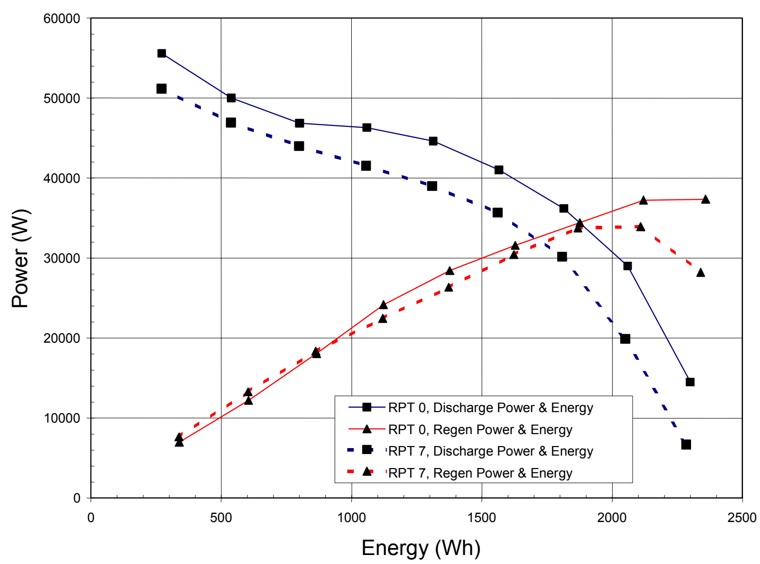

Figure 6. Saft Li-Ion Pulse Power vs. Energy Removed at Beginning of Testing and after 190,000 Cycles

To obtain the PNGV Power Assist available energy as a function of power, first the axes in Figure 6 are rotated such that the *Energy* becomes the dependent variable and *Power* becomes the independent variable. Then, the difference in energy between the discharge power curve and the regen power curve is calculated. This difference is defined as the PNGV available energy and is given by

$$E_{avail}(P) = E\ (P_{dis}) - E\ (P_{reg})$$

where $E(P_{dis})$ and $E(P_{reg})$ are the energies associated with P_{dis} and P_{reg}, respectively, after the axes rotation. The available energy as a function of power for this example is shown in Figure 7, again with a BSF of 44.3. [Note that the PNGV definition of available energy for the Dual Mode application is defined differently. For Dual Mode only, available energy is defined as the total energy released during a constant 6-kW discharge over the DOD range where the PNGV power goals can be met.]

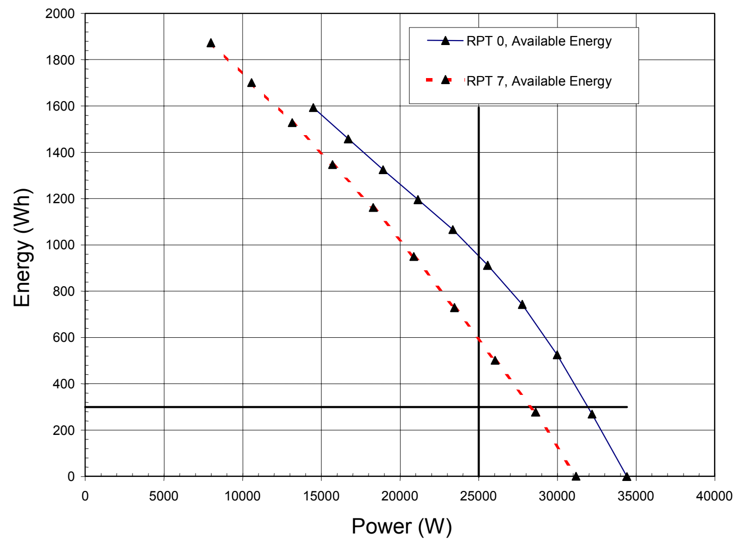

Figure 7. Saft Li-Ion Cell Available Energy vs. Discharge Power at Beginning of Testing and after 190,000 Cycles

Also shown in Figure 7 are bold lines indicating the PNGV Power Assist energy goal of 300 Wh and the discharge pulse power goal of 25 kW. As the cell ages, the available energy decreases and the curves shift to the left. As long as the cell's available energy curves stay to the right of the crossover of the two goal lines, the cell is able to simultaneously meet the PNGV energy and power goals. Conversely, if the cell's available energy curve had moved to the left of this crossover point, the cell would no longer have met the goals and testing would have been stopped. For this example, after 190,000 cycles the cell is still well able to meet the power and energy goals, and linear extrapolation indicates that the cell will likely meet the PNGV Power Assist 300,000 cycle requirement, as well.

As a further example, performance data from a full-size 6-Ah, 280-V, lithium-ion Saft pack that is nearing completion of cycle-life testing at INEEL are shown in Figure 8. The Saft pack is comprised of six modules, each containing twelve Saft HP-6 cells. The combination of these six modules with its associated hardware (the casing and the electronic control and thermal management systems) is generically referred to as a pack. By definition, the BSF for a full-size battery pack is 1.0.

Characterization testing was begun on the Saft pack at the INEEL in September 2000. It then began cycle-life testing at 30% DOD and 30°C, and to-date has successfully completed over 250,000 25-Wh Power Assist life cycles. (A sister Saft pack cycled at 30% DOD and 40°C successfully completed 300,000 cycles in November 2000.) These devices represent the maturest PNGV HEV battery technology.

At the beginning of cycling, the pack's PNGV pulse discharge power was about 32.6 kW and the available energy was about 710 Wh. And as shown in Figure 8, after completion of 220,000 cycles, the Saft pack was still well able to meet PNGV power and energy goals. At this point, the pack was able to provide 27.7 kW of discharge pulse power at the 300 Wh energy line and to provide 500 Wh of available energy at the 25 kW line. Also, throughout testing, the pack maintained an energy efficiency around 95%. This pack is projected to complete 300,000 cycles around March 2002 while continuing to meet the PNGV power and energy goals.

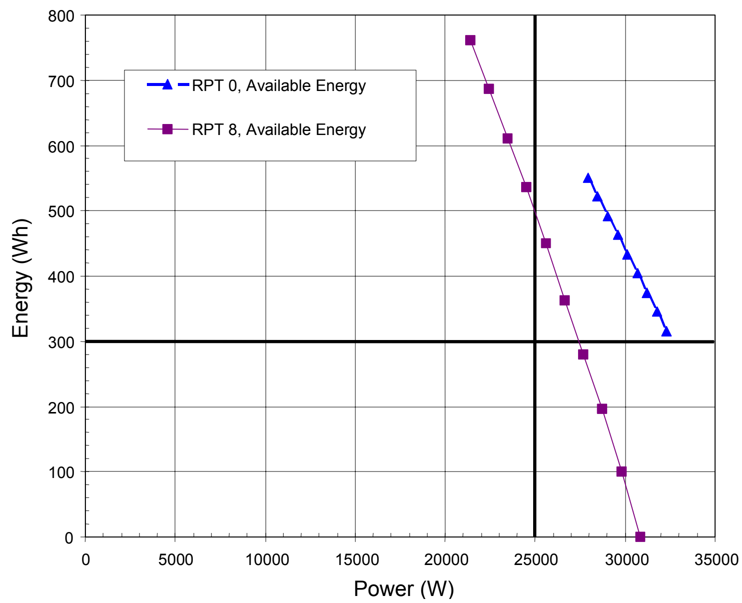

Figure 8. Saft Li-Ion Pack Available Energy vs. Discharge Power at Beginning of Testing and after 220,000 Cycles

Other new tools and methodologies are also being developed and utilized at the national laboratories to investigate degradation mechanisms that may impact cell life. For example, Figure 9 shows EIS Nyquist plots at four-week intervals during cycle-life testing for a representative lithium-ion cell from the DOE's Advanced Technology Development (ATD) Program (Ref. 4). This cell was cycled at 45°C and 60% SOC for 32 weeks at INEEL using the PNGV 25-Wh Power Assist Cycle-Life Profile. The BSF for this 18650-size cell is 553. To-date, the cell has accumulated over 270,000 cycles. The majority of the impedance growth in the curves lies in the frequency band between about 200 Hz to the trough frequency. The BOL trough frequency was about 2.5 Hz and has monotonically decreased to about 0.5 Hz at the end of 32-weeks of testing. Increases in the real impedance as the cell ages are related to growth of the thin film solid electrolyte interface (SEI) layer on the anode and/or cathode.

Lastly, a new measure of cell degradation under evaluation at the national laboratories is differential capacity, $\boldsymbol{Q_{dif}}$, (Ref. 5 and 6). It is given by

$$\boldsymbol{Q_{dif} = (1/Q)[d(Ah)/dV]}$$

where $\boldsymbol{Q}$ is the BOL capacity and $\boldsymbol{d(Ah)/dV}$ is the derivative of the capacity with respect to the voltage. Figure 10 shows a typical plot of differential capacity versus cell voltage calculated from a $C_1/25$ discharge and charge test for a representative ATD lithium-ion cell cycled at 45°C for 32 weeks at INEEL. Peaks are believed to be related to specific intercalation sites within the anode and/or cathode. The integrated area under each curve is equal to the BOL-normalized capacity of the cell. Thus, a decrease in the amplitude of a peak indicates that the cell's capacity has decreased over that respective voltage interval. It has been postulated that the degradation of cell performance with aging is related to both the changes in the amplitude and the location of these peaks. These changes may be a result of disruptions in the cathode crystalline lattice with aging.

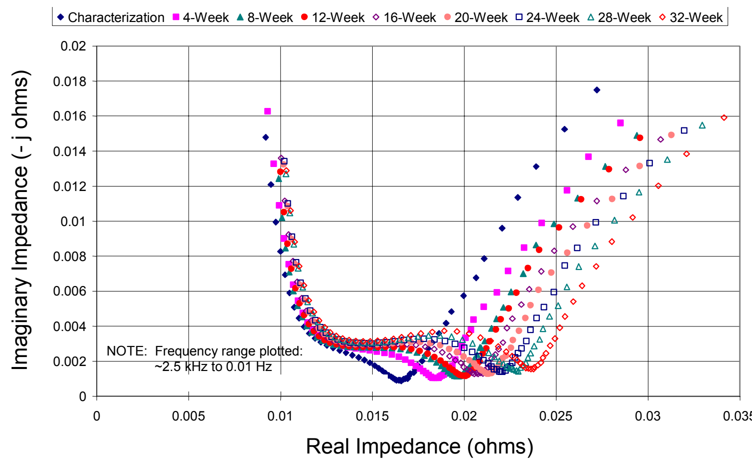

Figure 9. EIS for an ATD Li-Ion Cell over 32 Weeks of Life Cycling

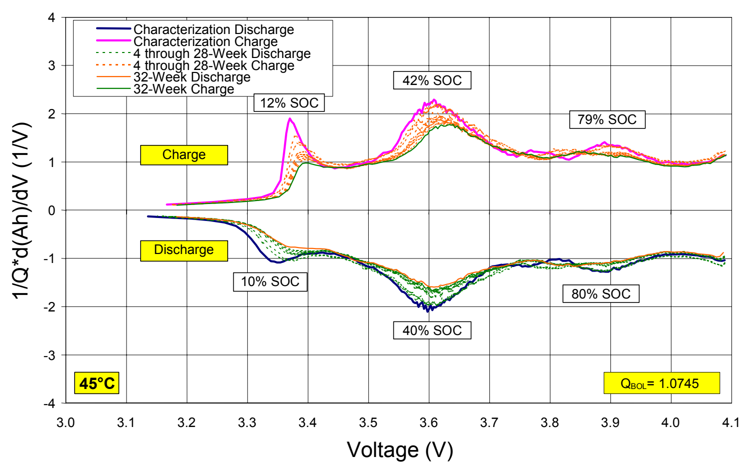

Figure 10. Differential Capacity Peaks for Li-Ion Cells Decrease During Life Cycling

LIFE MODELING

Cell degradation as a function of calendar time or cycle count and other test conditions is being investigated at several national laboratories. From either the HPPC data collected during the RPT's or from the pulse data during calendar- and cycle-life testing, discharge and regen resistances can be calculated as a function of time and test conditions. This information is being utilized to develop predictive life models for PNGV. Two distinct modeling approaches are being developed and evaluated by INEEL.

The first modeling approach is based upon the calculation of power fade over time as determined from the RPT's and associated available energy curves. Another set of six Saft 12-Ah lithium-ion HP-12 cells (1999 configuration) has been under test at INEEL for over 92 weeks using the PNGV calendar-life test. Two cells each are being subjected to temperatures of 40°C, 50°C, or 60°C. First, power fade as a function of time is calculated for each pair of cells at the three temperatures. This information can be used to construct an Arrhenius relation as shown in Figure 11, which enables extrapolation from the higher accelerated-aging temperatures to 25°C. The graph plots the natural logarithm of the "Years to End of Life" versus the inverse temperature in Kelvin and shows a projected calendar life of 16.5 years after 92 weeks of testing (Ref. 7). Testing is continuing and recent data show that the rate of degradation is decreasing, thus the calendar life of these cells may even be longer. In any case, battery developers are continuing their efforts to meet the PNGV calendar-life goal of 15 years, as well as other performance and cost goals.

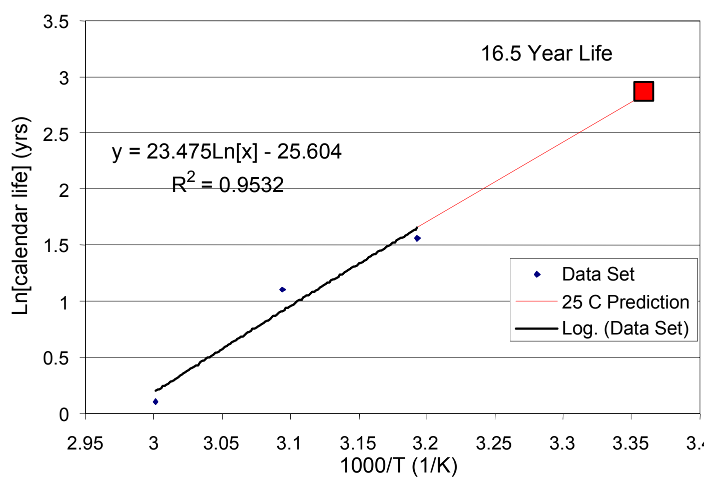

Figure 11. Calendar-Life Model of Saft HP-12 Li-Ion Cells

Through participation in the ATD Program, INEEL has also developed a second modeling approach for both calendar life and cycle life. For example, a calendar-life model was developed to account for the time, temperature, and SOC of the batteries during testing (Ref. 5). The functional form of the model is given by:

$$R(t,T,SOC) = a\{exp[b/T]\}t^{1/2} + c\{exp[d/T]\}$$

where $\boldsymbol{a}$, $\boldsymbol{b}$, $\boldsymbol{c}$, and $\boldsymbol{d}$ are functions of SOC, and where $\boldsymbol{b}$ and $\boldsymbol{d}$ are related to activation energies, $\boldsymbol{E_b}$ and $\boldsymbol{E_d}$, such that $\boldsymbol{b = E_b/R}$ and $\boldsymbol{d = E_d/R}$, and where $\boldsymbol{R}$ is the universal gas constant. (A similar approach has also been used to develop ATD cycle-life models (Ref. 6).)

The square-root-of-time dependence can be explained by either a one-dimensional diffusion type of mechanism, presumably of the lithium ions, or by a parabolic growth mechanism of a thin-film SEI layer on the anode and/or cathode. A diffusion type of mechanism would arise from the diffusion of lithium ions into or out of the electrodes, through the electrolyte, through the separator, or through the SEI layer. The thickness of the SEI layer is believed to increase with aging and hence increases the cell's electrical resistance.

Figure 12 shows a representative comparison of ATD calendar-life test results to the model at 80% SOC. The model fit is excellent at 40°C, 50°C and 60°C, but not at 70°C, where it is believed that a different physical mechanism is controlling.

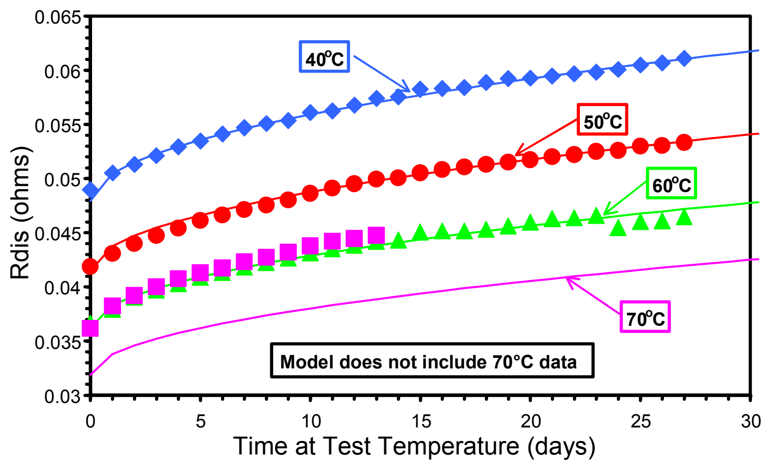

Figure 12. Calendar-Life Discharge Data and Model Predictions for ATD Cells at 80% SOC

As part of the ATD Program, ANL and SNL are also developing life models. ANL is developing resistance growth models and SNL is utilizing data from their accelerated life testing to develop power and capacity fade models. The SNL modeling approach is based upon both linear and nonlinear regression analyses. A report of the ANL work is found in Reference 8 and that of SNL is found in Reference 9.

Others also are involved in modeling as reported in the OAAT-sponsored Workshop on Development of Advanced Battery Engineering Models. The topics discussed at the Workshop covered fundamental physical phenomena, thermal models, performance and economic models, and vehicle and power system simulation models. The workshop concluded with a discussion of data needs and sources. A report of the workshop is found in Reference 10.

CONCLUSIONS

The DOE's OAAT and the PNGV are investigating and funding the development of advanced high-power batteries for HEV applications. Under their auspices, new PNGV testing procedures and analytical methodologies have been developed. These enable the testing of various chemistries, technologies, and sizes of products and provide objective comparison of results. Also, calendar-life and cycle-life models are under development and evaluation at the national laboratories that enable the extrapolation of accelerated-aging test data to normal operating conditions. Recent performance data for Li-ion cells and packs show that PNGV power and energy cycle goals can be met and that the calendar life is about 15 years. Lastly, the national laboratories are continually exploring new testing and analytical methodologies to further aid the OAAT and PNGV to understand fundamental electrochemical degradation processes and to overcome technical barriers to the commercialization of lithium-ion batteries for HEV's. This is expected to continue under FreedomCAR.

ACKNOWLEDGEMENTS

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